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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Evolution of aerosol optical thickness over Europe during the August 2003 heat wave as seen from CHIMERE model simulations and POLDER data" *by* A. Hodzic et al.

A. Hodzic et al.

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The authors are thankful to both reviewers for their work. According to their rapports, both reviewers are in favor of publication of this paper in Atmospheric Chemistry and Physics although they raise a number of important questions addressed in this revised version of the manuscript.

1. Specific Answers to Reviewer 1

General comment from Reviewer 1: *This paper describes an interesting comparison of atmospheric aerosol measurements by the POLDER satellite instrument and the surface network, with the CHIMERE regional-scale model. The episode chosen, the 2003 heat wave, is a key event for air-quality model evaluation and has the additional advantage of an improved coverage of the satellite measurements due to the absence of clouds. CHIMERE is a state of-the-art aerosol model and the POLDER instruments are dedicated to measuring aerosols. The comparison is presented well and provides considerable insight into the shortcomings of the model and measurements. I am in favor of publication in ACP.*

Comment 1: *Did the authors consider comparing with other existing satellite aerosol data sets? For instance the MODIS AOT data is available, and MODIS is already referred to in the context of the Portugal fires. Adding such a MODIS aerosol image (if this can be easily done) would give the reader a good impression on how well satellite retrievals agree, helping in the interpretation of the results. Or alternatively: motivate why the focus is on POLDER only. Why is the focus on the smallest aerosol particles?*

Answer 1: In our study only the comparison with POLDER data has been considered motivated by following reasons:

- At the time of our study we disposed of high-quality POLDER data for the comparison of AOTs with the model over Europe provided in the frame of our collaboration with LOA (Laboratoire d'Optique Atmospherique) colleagues. These data are well adapted to questions we raise here: the evolution and the origin of the atmospheric aerosol load encountered over Europe during the 2003 summer heat wave. As demonstrated in the manuscript, the large AOTs observed during this period are due to both anthropogenic aerosols produced locally and the advection of soot particles originating form Portuguese forest fires. Therefore, the use of the POLDER instrument for the model evaluation is a good choice as it is known to be very sensitive to aerosols over land surfaces and particularly to the smallest aerosol fraction that is mainly issued from anthropogenic sources and biomass burning. Our choice is explained in the manuscript on pages 3-4.

- As suggested by reviewer, it is interesting to consider data from another sensor in order to complete our analysis. The following figure shows the comparison between MODIS and POLDER AOT retrievals during 4 and 5 August. We can see that MODIS AOTs (at 550 nm) display similar spatial structures as POLDER data on 4-5 August and confirm the presence of an aerosol layer over the Northern Europe. This figure (Figure 2b) has been added to the new version of the manuscript in order to strengthen our results. However, we do not focus on the analysis of MODIS data in our comparison as the MODIS AOT values are not directly comparables with POLDER ones: MODIS AOTs include whole aerosol distribution and are made at 550 nm, while POLDER AOTs account only for fine-mode aerosols at 865 nm.

We added a new figure (Figure 2b) and the following comment in the new version of the manuscript:

- Figure 2b: Geographic distribution of the aerosol optical thickness at 550 nm retrieved from the MODIS sensor over Europe on 04 and 05 August 2003. MODIS data account for both fine and coarse mode aerosols and are represented with 0.25x0.25 degree resolution.
- The comparison with MODIS data shown on Figure 2b also confirms the presence of high AOT values (> 0.9 at 550 nm) over the Northern Europe on 4-5 August. These results indicate that a particular process, which is not included in the model formulation, may be present in reality.

Comment 2: *The authors discuss long-range transport aspects. Please provide a short discussion to convince the reader that a top level at 500 hPa is sufficient to* *describe free troposphere tracer transport, or mention possible problems due to this 500 hPa upper level. The lidar measurements suggest that there is still a considerable aerosol signal at about 5km.*

Answer 2: We agree that a top level of 500 hPa may be a limiting factor when studying long-range transport of smoke particles as these particles can be injected at higher altitudes. As already discussed in the manuscript (sect. 4.6), the altitude of the fire emission injection is a key factor influencing the transport of smoke particles. Therefore, in order to answer the reviewer's question and to quantify the impact of the model top level limitation on the long range transport of particles we performed an additional run with increased model top level (up to 300 hPa). As shown on figure 2a, the long-range transport of particles is not significantly influenced by the model top altitude during our study case (5-6 August) as the particles propagate at the altitude of 3-5 km. However, the thickness of the smoke layer is probably underestimated. In this study the underestimation of the aerosol layer thickness is not a limiting factor as we do not calculate the associated AOT and we are only interested in its origin. However, we agree that studying the transcontinental transport of pollutants or including the wild fire emissions in further model versions will require increasing the model top level.

This is now explained in the revised version of the manuscript: *Finally, it is important to notice that a model upper boundary of 500 hPa could be a limiting factor when studying free tropospheric transport of smoke particles since these particles can be convected to much higher altitudes. Additional sensitivity studies regarding to the model's top level altitude revealed that the long-range transport of particles is not significantly changed during our specific case study by increasing the model top altitude. However, considering the real wild fire emissions in further model studies will require increasing the model top level at least up to 300 hPa.*

Comment 3: *The lidar measurements show an interesting development of aerosol over the 24 hour period with a pronounced peak at 18:00. Such detailed measurements ask for a more detailed analysis. It would be quite interesting to see how well the tracer*

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version is able to capture both the timing and vertical distribution. Is it possible to produce a plot of the 24 hour period over Cabauw, to be added as a panel 6b in the paper?

Answer 3: The lidar measurements show the evolution of the vertical structure of the lower troposphere over the 24 hour period. However, it is not possible to reproduce the equivalent plot for model simulations as it requires several assumptions on the tracer's chemical composition. The evaluation of the model ability to simulate the aerosol load within the PBL has already been demonstrated in previous studies (Hodzic et al., 2004) and this study is focused on the evaluation of the spatial correlation of integrated aerosol concentrations provided by the model and measurements. The plot of the evolution of the vertical distribution of tracers' arbitrary concentrations is not really relevant and is not directly comparable to the lidar plot. Therefore we decided not to include it in the new version of the manuscript.

Comment 4: *On page 4117, l 9-20, the authors make a strong statement about model formulations which are not well suited for extreme weather conditions. Such a statement asks for further discussion in the paper, but such a discussion is missing (the discussion section could be a good place). Are the authors able to estimate how these aspects influence the aerosol levels modelled by CHIMERE during the heat wave?*

Answer 4: Model parameterisations have rarely been tested in the case of extreme weather conditions and are subject to large uncertainties. The main difficulties are already explained on page 4117 and concern mainly the biogenic emissions, as well as the aerosol emissions due to sporadic sources such as forest fires:

Modeling such a wide pollution episode is a challenging problem because models have to deal with an exceptional environment for which their parameterizations are not necessarily appropriate. For instance the formulation of classical models of dry gaseous deposition or biogenic emissions do not generally account for the exceptional deficit in soil water. When using standard anthropogenic emission inventories chemistry-

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transport models (CTMs) probably underestimate the evaporation of anthropogenic non-methane volatile organic compounds due to extremely high temperatures. For aerosols the drought increases the erodibility of soils and favors forest fires, phenomena which are generally not taken into account in an accurate manner in these standard emissions. Therefore in such extreme weather conditions there is a large degree of uncertainty in aerosol sources and physical parameterizations, leading to significant difficulties for CTMs to simulate the aerosol distribution and there is a strong need for the models to be tested against observations in such cases.

At this stage we are not able to provide an accurate evaluation of the degree of uncertainties in aerosol sources. But, the parameterization of fire emissions sources and their transport are a part of our future work in collaboration with the NCAR colleagues.

Moreover, the aerosol re-suspension processes could also play an important role during dry soil conditions and modify considerably the aerosol size distribution (Vautard et al., 2005). These processes are not included in the current model version.

Comment 5: *On page 4120 the POLDER retrieval is discussed, and an accuracy of 20-30% is claimed. This seems to be in conflict with the large differences found between POLDER and AERONET in sec. 4.4. Please comment. Does the 20-30% number need adjustment? How is this 20-30% estimate obtained?*

Answer 5: We agree that the difference between POLDER accuracy of 20-30% and the large discrepancies with AERONET data could be confusing. The accuracy of 20-30% on POLDER retrievals is issued from previous validation studies carried out by Deuze and his collaborators. Indeed, the POLDER-2 validation exercise was based on all AERONET sites that were operational during the validation period. Considering the AERONET dataset with an Angstrom exponent greater than 1.5 (additional criteria) and rejecting desert regions, the global error on the POLDER AOT was found to be within 30%. During that validation exercise, POLDER-2 AOTs have been compared with fine-mode AERONET AOTs which are recomputed from the retrieved size distri-

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bution (integration stopped at Rmax=0.6 micron). It has been shown that the same computation done for Rmax 0.5 micron gave better agreement with POLDER-2 AOT on the available dataset.

Besides, the POLDER algorithm was shown to be more efficient when the polarized aerosol signal is important and when the ground contribution is rather low. In the present study, only European sites are considered (fig. 5) and most of them are characterized by low AOT. Therefore, as we already explained, the discrepancies could reach 100% and more for small aerosol amount over land surfaces. It is also know from recent studies that MODIS retrievals over land exhibit the same failure.

This is now explained more precisely in the new version of the manuscript: *A lognormal size distribution is assumed and the aerosol refractive index is fixed to m=1.47-0.01i based on climatology (i.e, single scattering albedo ranging from 0.85 to 0.95 depending on the size distribution shape). Over land surfaces (except for desert regions), considering all AERONET sites available during the global validation exercise, departure between satellite and ground-based accumulation aerosol optical thickness has been shown to be less than 20-30% when medium and high anthropogenic or biomass burning aerosol loadings are considered. Discrepancies could reach 100% and more when small AOT occurs simultaneously with inaccurate surface modelling.*

Specific remark: Past and present tense is mixed in the paper. Please check that present tense is used everywhere.

Answer: English has been improved in this new version of the manuscript.

2. Specific Answers to Reviewer 2

General comment from Reviewer 2: *This paper presents an interesting case study of the modelled and derived aerosol optical depths over Europe during the heat wave*

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of 2003. I find the results interesting and an appropriate subject for ACP, and would recommend publication after the points raised below are addressed.

Comment 1: *While the paper points out that the POLDER retrievals appear to have a low bias with respect to the observations (primarily AERONET), there is little quantitative assessment for why the errors in the retrievals exist. Indeed, p4120, l10-15 suggests that over land the biases should be less than 20-30% in 'medium and high' aerosol optical depth conditions. Subsequently (P4126) the authors point at three potential sources of errors, but do not investigate which (if any) of these sources of error are the most likely culprits. With regard to the aerosol model, it would be worthwhile modifying the POLDER retrieval with (for example) a modification to the aerosol absorption to see what single scattering albedo would be needed to bring the results into agreement with the AERONET data. Similarly, the size distribution assumed in the POLDER retrievals could be varied. In this way the shortcomings of the POLDER retrieval could be systematically assessed. I would suggest that the data from AERONET be used in these retrieval algorithms as a starting point.*

Answer 1: As already explained in the manuscript, the quality of satellite retrievals depends on the aerosol amount and the surface conditions. A low accuracy is encountered in case of: - small AOT (same for all sensors), - big failure in the surface model (over urban or desert region). The same error occurs for MODIS sensor, with Kaufman relationships respectively between 2.2 and 0.49 at 0.67 microns, - big and/or non spherical particles (this is also problematic for MODIS).

When aerosols optical thickness is small (i.e. less than 0.1 at 870 nm), wrong surface model can yield an underestimation (about a factor 2) on AOT. In these situations, the error mainly comes from the surface correction. This kind of error vanishes when AOT increased, as shown by data obtained during the wild fire episode plotted on figure Fig.5. In addition, absorption is taken into account in the aerosol model used in the retrieval (imaginary part is 0.01 which correspond to single scattering albedo ranging from 0.85 to 0.95 depending on the size distribution shape). Potential error due to absorption assumption is far from the observed departure between POLDER and AERONET.

This is now explained more precisely in the new version of the manuscript:

The underestimation of the POLDER AOT at 865nm is consistent with previous POLDER validation studies (Deuzé et al., 2001) and can be explained by large uncertainties in the aerosol retrievals from satellite data. The satellite underestimation could be associated to numerous factors such as (i) uncertainties in the surface polarization correction, (ii) underestimation in the detection of fine fraction of dust particles, or (iii) the incorrect choice of the aerosol model used for the retrieval of aerosol optical properties. In this study, the main error comes from inaccuracies in the surface modelling that reaches its maximum over urban surfaces. Simultaneously, background AOT is generally not very high in the considered area. Combination of these two factors yields to strong underestimation of AOT (Deuzé et al. (2001) and Nadal and Bréon (1999)). Another potential error is related to the occurrence of low polarizing particles (dust particles) that also affects the retrieval accuracy. However, the impact of this latter error is limited regarding the geographical location (Europe) and the studied period.

Comment 2: *Regarding the modelling, an arbitrary amplitude passive tracer is added to account for the fires. Why not just add emissions of organic/black carbon/inorganic components to account for the changes in the optical depth. The authors could iteratively adjust the emission per square metre until agreement between the model and the observation was reached. It would be interesting to see whether the emissions in terms of particle mass per square metre could be reasonably estimated by this technique.*

Answer 2: The reviewer suggestion is a very interesting point, certainly an exciting future work to do with our model. To achieve a real study of forest fires impact on the air quality over Europe, many processes are required. At this time, these processes are not well known and need an important step forward in measurements and modelling (how to model hot convective vertical plume induced by fires? How to parameterize the

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organic/black carbon/inorganic mass as a function of the landuse, the soil humidity, the temperature etc.?). These aspects are beyond the scope of our paper: in the present study, we just want to check if one (and just one) particular plume observed over The Netherlands may be originated from Portugal. This is why we choose only one passive tracer emitted in Portugal: using this approach we are able to follow one plume (and this approach avoids mixing between several plumes). Our work is not to implement a realistic landuse scheme of fires (not yet but certainly in the next years), but to follow one very specific and isolated event. We thus retain the interesting idea of the reviewer for a future work.

Specific remarks: P4126. Three potential areas of error are highlighted, but not investigated further. Given that AERONET provides some data on the aerosol size distribution and the refractive index, it would be worth modifying the POLDER algorithm to see if this source of error can explain the differences. P4130, l12-15. With some more effort the reasons for the differences between the model and the POLDER observations wrt AERONET could be elucidated. In this way, both the model and the POLDER retrievals could be improved. The paper currently only points out that there are differences, and stops short of suggesting why these differences occur (for POLDER) and short of quantifying the emission of biomass burning aerosols necessary to bring the model into line with the observations.

Answer to specific remarks: For our study we use high quality POLDER AOT products provided by LOA colleagues. Unfortunately, modifying the POLDER retrieval algorithm is beyond our competences and can not be done in this study. Moreover, we agree that this study constitutes a preliminary work that highlighted the major discrepancies between regional model simulations and satellite data, and identified the missing processes that need to be improved/included in aerosol models. Our further work will focus on the introduction of real fire emissions and associated parameterizations in order to bring the model simulations in line with satellite observations.

Please correct the following figure captions:

- Figure 4: Time correlations between simulated and POLDER retrieved AOT across Europe during the summer 2003 heat wave (from 1 to 15 August 2003).

- Figure 8: Transport of smoke particles from forest fires in Portugal through Europe as simulated by CHIMERE model. Vertical profiles of boundary layer (BL) and free troposphere (FT) emitted tracers (a.u: arbitrary unit) are presented at different locations.

Interactive comment on Atmos. Chem. Phys. Discuss., 5, 4115, 2005.